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Carboxymethyl Cellulose From Palm Oil Empty Fruit Bunch – Their Properties And Use As A Film Coating Agent.

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ABSTRACT

This study was carried out to determine the physicochemical properties of carboxymethyl cellulose (CMC) derived from cellulose of palm oil empty fruit bunch (EFB) and its use asa film-coating agent. Samples were prepared at various concentrations and then their physicochemical properties were studied including the viscosity, pH, tensile strength of films, surface properties of the films and dissolution studies on coated tablets. CMC EFB showed lower viscosity than commercial CMC product at the concentration of 1%, 2% and 3% with the values of 44.0cp, 299.9cp, 358.9cp and 90.0cp, 689.9cp, 5569.0cp respectively. The tensile strength of the films for CMC EFB were 7.85MPa, 14.79MPa, 10.36MPa while the commercial CMC exhibited higher values of 21.72MPa, 35.14MPa and 26.9MPa at similar concentration. The scanning electron microscope showed different surface properties of the films for both of them where the commercial CMC is smoother in texture and very transparent unlike its counterpart. However, dissolution studies on paracetamol tablets coated using the samples showed no significant difference ($p>0.05$) in drug release profile between the two materials. Hence, CMC EFB has a greater potential to be developed as a competitive tablet-coating agent despite the differences in its physicochemical properties.

Keywords: carboxymethyl cellulose, palm oil empty fruit bunch, tensile strength, scanning electron microscope, tablet coating.

ABSTRAK

Kajian ini dilakukan untuk menentukan sifat-sifat fizikokimia carboxymethyl cellulose (CMC) diperolehi dari selulosa tandan kosong buah kelapa sawit (EFB) dan penggunaannya sebagai ejen penyalut tablet. Sampel disediakan dari pelbagai kepekatan dan sifat-sifat fizikokimia dikaji termasuk dari sudut kelikatan larutan, pH, kekuatan ketegangan filem yang terbentuk, sifat permukaan filem dan ujian pelarutan tablet bersalut. Hasil kajian menunjukkan perbezaan sifat-sifat fizikokimia di antara CMC dari EFB berbanding dengan sediaan yang komersil. Kelikatan CMC EFB pada kepekatan 1%, 2% dan 3% adalah 44.0cp, 299.9cp dan 358.9cp berbanding sediaan komersil iaitu 90.0cp, 689.9cp dan 5569.0cp pada kepekatan yang sama. Kekuatan ketegangan filem CMC EFB pula mendapati nilai yang rendah iaitu 7.85MPa, 14.79MPa dan 10.36MPa berbeza dengan produk komersil iaitu 21.72MPa, 35.14MPa dan 26.9MPa. Ujian imbasan mikroskop elektron menunjukkan perbezaan permukaan yang ketara di mana

filem CMC komersil kelihatan lebih halus dan telus berbanding CMC EFB. Namun begitu, ujian pelarutan tablet paracetamol bersalut tidak menunjukkan perbezaan yang signifikan ($p>0.05$) terhadap profil pelepasan dadah untuk kedua-dua sediaan. Ini menunjukkan bahawa CMC EFB mempunyai potensi untuk dibangunkan sebagai ejen penyalut tablet walaupun terdapat perbezaan sifat-sifat fizikokimia.

Kata kunci: carboxymethyl cellulose, tandan kosong buah kelapa sawit, ketegangan, imbasan mikroskop elektron, penyalutan tablet.

INTRODUCTION

Palm oil industry in Malaysia is one of the most important key sectors that generate the economic growth. However, the production of large amount of biomass waste accompanying the industry is becoming a major concern. It has been estimated that, in the year 2000 alone, 3.08 million tones of empty fruit bunch (EFB) was produced based on 42.5 million tones dry weight of fresh fruit bunch (Kamaruddin et al. 1997). The EFB, consists of 60% of cellulose and hemicellulose, and their use for the production of value-added products is highly recommended. (Abdul Azis et al. 1989 and Rosnah et al. 2000)

In general, the primary cell wall of plants contains 10-20% of cellulose and the secondary cell wall consists up to 50% cellulose. These compositions however, differ from one plant to another and vary with age. The cellulose exists together with structures like lignin and hemicellulose and is stabilized with the presence of hydrogen and ionic bondings (Allan 1995). The type and design of the cellulose determine the characteristics of the mechanical fibre and plant tissues. It also relies on the chemical long chain basic structure of the cellulose, which is based on the number of glucose units or cellobiose. The degree of polymerization of the glucose units will also be taken into account in order to determine the grade and the strength of the mechanical fibre obtained from the tissue plants. On the other hand, one unit of cellobiose is made of two glucose molecules (*D-glucopyranoside*), which combines together at position β (1→4). This combination will form a long chain chemical unit branching and eventually determines the degree of polymerization of the cellulose (Solomons and Fryhle 1976).

Cellulose and its derivatives have been used widely in pharmaceutical industry especially as a tablet film-coating agent (Aulton 1988). These include hydroxypropyl methylcellulose, hydroxypropyl cellulose, sodium carboxymethyl cellulose and hydroxyethyl cellulose.

There are many attempts to diversify the use of lignocellulosic derived from EFB (Rosnah et al. 2000). One of the products that are receiving major attention is the production of carboxymethyl cellulose (CMC) mainly for the pharmaceutical industry such as a tablet film coating agent and gelatin substitute for capsule (Aulton 1988). CMC has been known as a water-soluble polymer used mainly for tablet coating either as a non-functional coating, sustained or controlled release preparations (Banker and Rhodes 1996). Literature reviews have shown that similar studies on EFB have not been carried out. Hence, the objectives of this study are to investigate several physicochemical and film-coating properties of the material and compared with the commercial CMC.

Understanding the properties and characteristics of this crude material will help in enhancing its quality for pharmaceutical uses.

MATERIALS AND METHODS

pH AND VISCOSITY MEASUREMENTS

Carboxymethyl cellulose (CMC) from empty fruit bunch (EFB) (Malaysian Palm Oil Board) and commercial CMC (Sigma Chemical Corp. USA) solutions were prepared at three different concentrations namely at 1%, 2% and 3% by dissolving the materials in distilled water and heated at 60^{o}C. The solutions were cooled down before the pH measurements were made at room temperature using a pH meter (model MP225, Mettler-Toledo Ltd, England) inside a water bath to maintain a constant temperature. Viscosities of the solutions were also measured using a viscometer (model DV-11, Brookfield, USA) at room temperature using spindle no. 3.

FREE FILM PREPARATIONS

Twenty ml of each sample solutions were poured into individual Petri dish and balanced in such a way that it covered the whole surface of the dish. The procedures were performed with cautious in order to avoid any bubbles or contaminants from entering the dish. The Petri dishes were then kept in an oven for 24 hours at 45^{o}C. Once dried, the films were removed and kept in a dessicator at room temperature before further use.

TENSILE STRENGTH MEASUREMENT

The films were cut as required and tested using a Universal Testing Machine (Instron 5567) at a speed of 5.0 mm/min using 500N load cell. The thickness of the films was limited between 0.05-0.20 mm. Five readings were recorded for each sample.

SCANNING ELECTRON MICROSCOPE (S.E.M)

Cast films of CMC EFB and commercial CMC of different concentrations were scanned under electron microscope (model XL 30 FEG, Philips Corp, Holland) equipped with a scanner (model EDACS-CDU LEAP).

COATING OF TABLETS

Tablets containing 500mg paracetamol (Millidon, Malaysian Pharmaceutical Sdn Bhd) were coated manually using a special device made for this purpose. The tablets were dipped and removed from the solution several times and dried throughout the process using a hot air blower. The process was done with cautious in order to get a uniform coating. The thickness of the coating at several spots on the tablet was measured as well as the weight gained from the above process.

DISSOLUTION TESTS

Dissolution was carried out using a dissolution tester (model TDT-08L, Electrolab Company, India) Type II according to *United States Pharmacopoeia* (USP 2004). Phosphate buffer solution was used as the dissolution media and the tests were carried out at 37^{o}C. The pH of the media was maintained at 5.8. Samples were taken at every 5-minute interval for 45 minutes. The samples were tested for the presence of paracetamol concentration using a UV spectroscopy (model UV-1601, Shimadzu, Japan) at the wavelength of 243 nm. A calibration curve was earlier prepared in order to determine the drug concentration when the absorbance values were obtained. Phosphate buffer solutions were used to dissolve paracetamol at the concentrations of 0.01, 0.02, 0.03, 0.04 and 0.05g/ml before the absorbance values were measured.

RESULTS AND DISCUSSIONS

Table 1 shows the pH values for all concentrations of CMC EFB and commercial CMC solutions for tablet coating. The pH values increases slightly as the concentration of the solutions increased. However, the pH values between the materials did not differ very much. The pH values within this range are acceptable for oral use as reported by Duchene et al. (1988).

TABLE 1 pH values for all carboxymethyl cellulose solutions.

CONCENTRATION (%)	pH VALUES	
	COMMERCIAL CMC	CMC EFB
1%	7.22 ± 0.0095	7.23 ± 0.0052
2%	7.24 ± 0.0063	7.26 ± 0.0150
3%	7.45 ± 0.0098	7.57 ± 0.0098

Figs. 1 and 2 show the Scanning Electron Microscope (SEM) photomicrograph of 1% commercial CMC and 1% CMC films respectively. It can be seen that the surface of the commercial CMC film is smooth compared to more fibrous-look film of CMC EFB. This may be due to the crude process of obtaining the CMC from the EFB. Although the SEM

of the CMC EFB film is not as shining or smooth as commercial CMC, the texture can be improved by adding a plasticizer into the formulation. The addition of plasticizers into film coating agents is well reported and widely used (Donbrow & Friedman 1974, Fell et al. 1979, Leopold 1999).

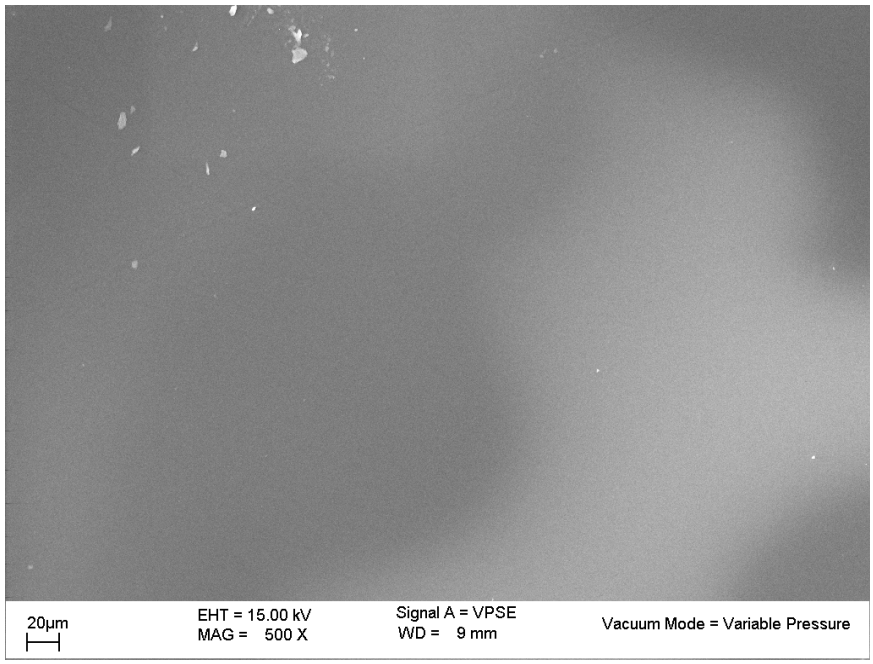

FIGURE 1 SEM photomicrograph of 1% commercial CMC film.

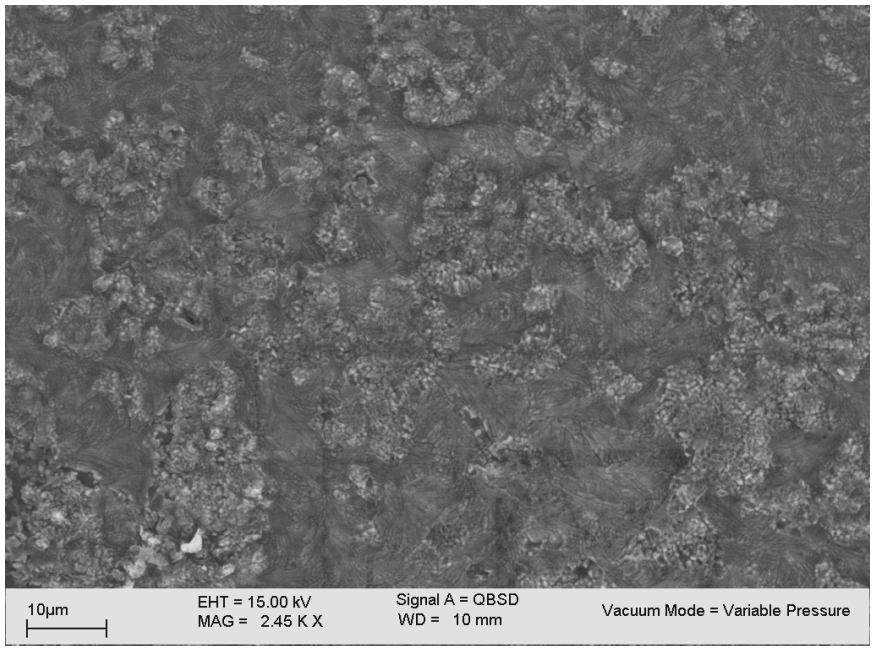

FIGURE 2 SEM photomicrograph of 1% CMC EFB film.

Fig. 3 shows the relationship between the viscosities of the solutions and concentrations for both materials. The results show that the viscosity of the commercial CMC exhibits a simple plastic flow rheology property. On the other hand, the viscosity of the CMC EFB increased slightly as the concentration increases. This suggests that the viscosity of the CMC EFB is more predictable compared to the commercial CMC. The rheological behaviour of the CMC EFB was probably mainly due to the presence of impurities during the extraction of the cellulose. The difference can also be seen very clearly from the scanning electron photomicrographs of the two samples. Although the measurements were only taken at three different concentrations, the behaviour of the polymers was in accordance with other researchers reported before (Banker et al. 1964).

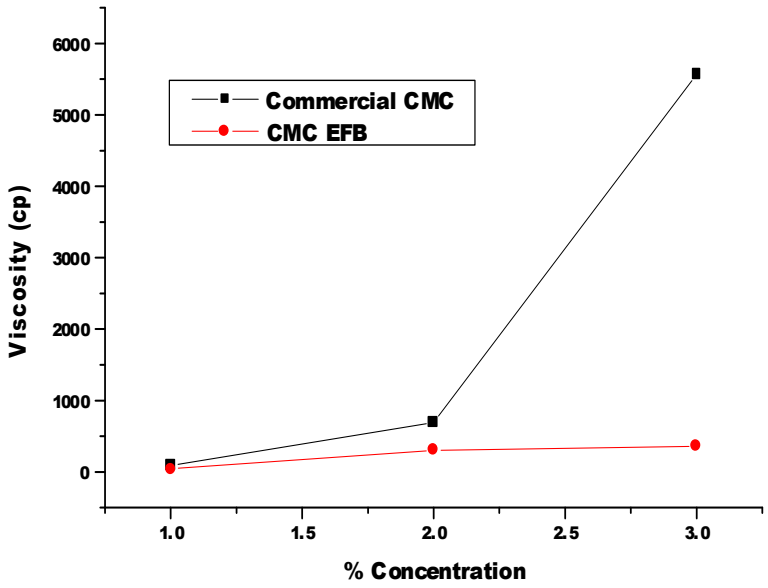

FIGURE 3 The relationship between the viscosities and the concentration (n=5).

Fig. 4 shows the relationship between the tensile strength of the films for both materials as the concentration increases. The result shows that the tensile strength of CMC EFB is lower than the commercial CMC. This is probably due to the presence of high fibrous materials in the films as shown in the SEM photomicrograph, which may affect the strength of the film. However, it can be noted that both materials exhibited a maximum tensile strength at 2% concentration. The decrease in the tensile strengths could be observed for both materials at 3% concentration.

Fig. 5 shows the calibration curve of the standard paracetamol concentration and its absorption at 243nm wavelength. Fig. 6 shows the relationship between the amounts

of paracetamol concentrations released against time. In general, the drug released was delayed from all the coated tablets compared to the uncoated ones. However, the tablets showed zero-order release as it reached 45 minutes of dissolution time. Comparing the results between the two materials showed no significant difference ($p>0.05$) on the drug release profiles. This suggests that, although there are some differences in terms of physicochemical properties between the two materials but as tablet coating is concern, the differences are perfectly fine.

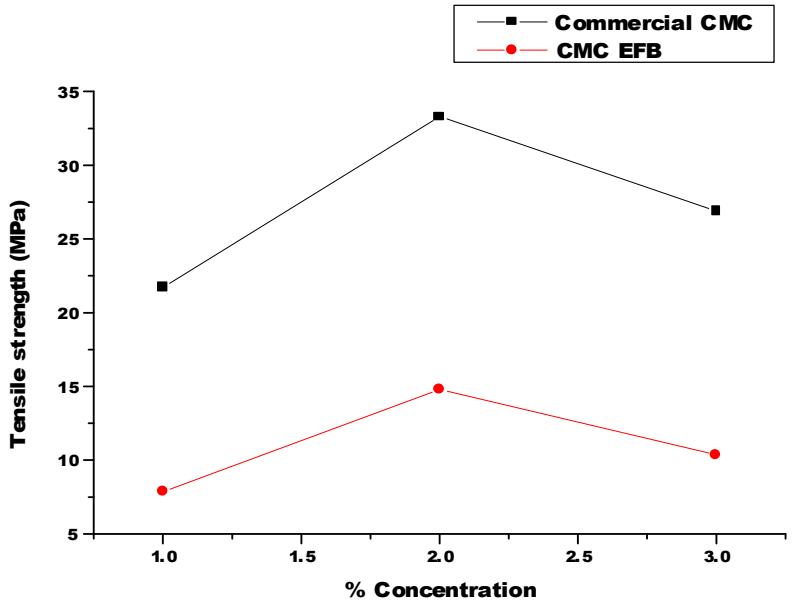

FIGURE 4 The relationship between the tensile strength and the concentration (n=5).

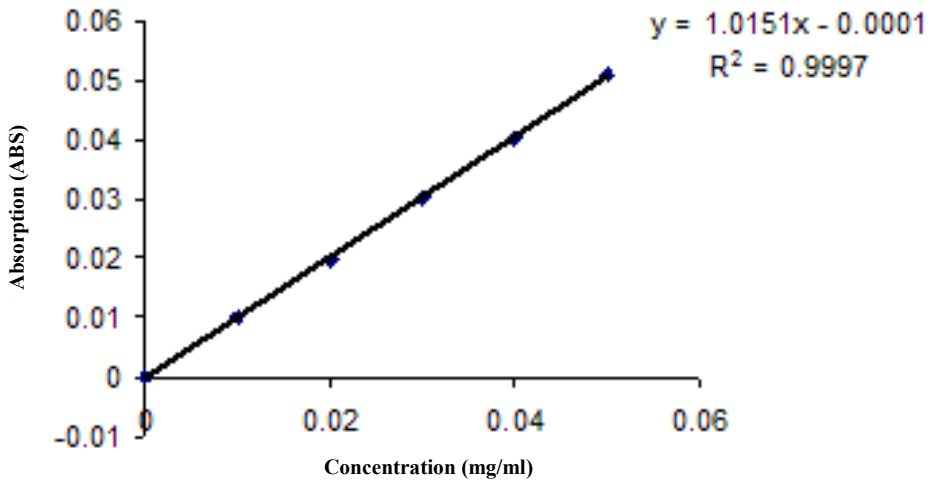

FIGURE 5 Calibration curve for standard paracetamol.

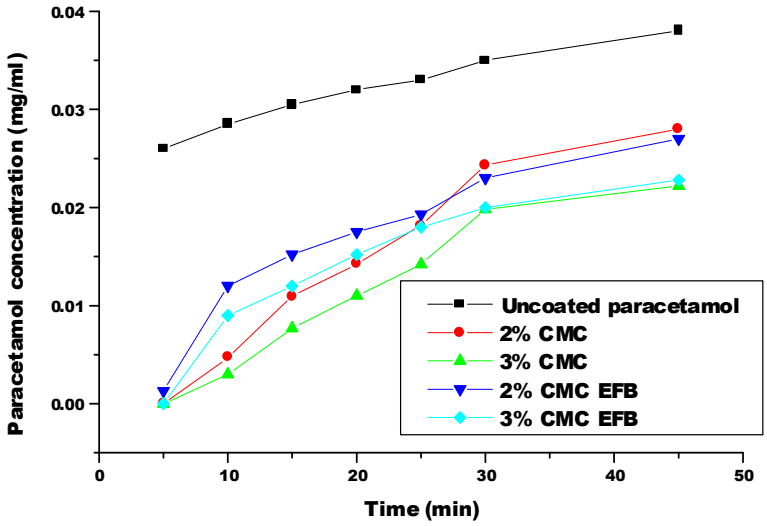

FIGURE 6 The relationship between the amounts of paracetamol released against time.

CONCLUSIONS

The results of the study suggested that the CMC EFB was incomparable to commercial CMC in terms of quality. However certain methods can be employed in order to improve the physicochemical properties of CMC EFB. This can be done by improving the method of extraction of the cellulose as well as by addition some plasticizers into the film formulations. The scanning electron photomicrograph showed that the films were more fibrous and the tensile strength of the films were shown to be weaker than the commercial CMC. However, the dissolution studies on the coated tablets suggest that CMC EFB has the potential to be developed as a tablet-coating agent despite the differences in the physicochemical properties of the two materials.

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